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16. ABSTRACT

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Increasing Cd additions to the soils, whether as $CdSO_4$ or in sewage sludge, resulted in increased Cd uptake by the plants, with the greatest accumulation of Cd by those plants grown on the acid soils. Liming the soils resulted in a decrease of Cd uptake by the two test crops. Regardless of soil pH, corn plants accumulated significantly higher concentrations of Cd than the Swiss chard plants. Results indicated that the added Cd was less available in the soils which had previously received sludge. In those pairs of soils which behaved differently, the pH of the soils which had previously received sludge was lower; thus, the added Cd would be expected to be more available. A comparison of the soils to which no $CdSO_4$ had been added indicated that the Cd originating from sludge was still available for plant uptake. The data indicate the need to consider previous sludge application (cumulative Cd additions) in evaluating potential Cd accumulation by plants from current (annual) Cd additions.

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CADMIUM SULFATE APPLICATION TO SLUDGE-AMENDED SOILS I. EFFECT ON YIELD AND CADMIUM AVAILABILITY TO PLANTS*

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ABSTRACT

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INTRODUCTION

In the past 15 years, knowledge of Cd in plants and soils has increased rapidly. Soil pH and organic matter are two soil factors that have been found

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TABLE 1

Sampling location and classification [13] of the soils used in the experiment

Soil series	Location	Family	Subgroups
Congaree	Macon, GA	Fine-loamy, mixed, nonacid, thermic	Typic Udifluvents
Salinas	Las Virgenes, CA	Fine-loamy, mixed, thermic	Calcic Pachic Haploxerolls
Britwater	Springfield, MO	Fine-loamy, mixed, mesic	Typic Paleudalphs
Burkhardt	Chippewa Falls, WI	Sandy, mixed, mesic	Typic Hapludolls
Pembroke	Hopkinsville, KY	Fine-silty, mixed, mesic	Ultic Paleudalfs
Russell	Frankfort, IN	Fine-silty, mixed, mesic	Typic Hapludalfs
Morley	Kendallville, IN	Fine, illitic, mesic	Typic Hapludalfs
Ross	Columbus, IN	Fine-loamy, mixed, mesic	Cumulic Hapludolls
Del Rey	Sandusky, OH	Fine, illitic, mesic	Aeric Ochraqualfs
Wea	Dayton, OH	Fine-loamy, mixed, mesic	Typic Argiudolls
Blount	Marion, IN	Fine, illitic, mesic	Aeric Ochraqualfs

to have a consistent effect on Cd uptake in nearly every crop. Uptake by plants of Cd has generally resulted in lower accumulation in neutral than in acid soils [1–5].

In greenhouse and field studies with sludge-applied Cd, liming the soils has generally reduced Cd uptake, but the amount of reduction depends upon the plant species and probably the soil as well. Studies by Chaney et al. [6] and Chaney and Hornick [7] showed a greater decrease in Cd uptake (80–90%) for limed plots in leafy and root vegetable crops than in oats and soybeans (0–70%) relative to unlimed plots. In a greenhouse study comparing four acid soils with four calcareous soils, Mahler et al. [3] found less Cd uptake by lettuce and Swiss chard plants grown on the calcareous soils.

Although plant accumulation of Cd has been found to be a function of many variables (soil pH, organic matter, cation exchange capacity, clay type and amount, hydrous metal oxides, carbonates and other inorganic compounds), it has generally been concluded that, for a system with constant soil, pH, and species, the Cd concentration in the plant exhibits a positive linear correlation with soil Cd addition. This concept is corroborated by the long-term land application sites studied by Otte and LaConde [8], Ryan [9], and Chaney and Hornick [7]. These studies indicate that the slope of the response curve is influenced by pH, crop and soil. Even when no Cd has been applied for a number of years, there is still a significant relationship between plant and soil Cd. This can also be deduced from studies by Mulla et al. [10] and Williams and David [11, 12] on their long-term phosphate plots. It appears that Cd remains available to plants once it is added to the soil, but that its relative availability may change with time.

Information on the relative availability of Cd from various land application practices and environmental conditions is essential to developing safe and beneficial ways of utilizing sewage sludge on land. This growth chamber study was undertaken to evaluate the effect of previous sludge additions to soils on the availability of inorganic Cd additions to soils and on yields.

TABLE 2

Chemical compositions of sludges for all sites [14]

Element	Range	Mean	Median
	Percent		
Vol. solids	36.1–74.7	51.1	52.4
NH_4-N	0.276–4.18	1.28	1.01
Organic-N	1.48–4.50	2.83	2.46
P	1.23–3.55	1.72	1.56
K	0.131–0.535	0.283	0.222
Na	0.195–0.798	0.481	0.611
Ca	1.39–6.25	3.53	2.78
Mg	0.359–1.23	0.700	0.585
Cl	0.25–1.09	0.36	0.275
H_2O	85.6–98.3	93.0	93.8
	$\mu g\ g^{-1}$		
NO_3-N	0.41–60.0	19.3	13.0
SO_4	8.6–348	95.4	81.7
Ag	0.95–10.2	3.21	1.95
As	1.13–6.21	3.08	2.52
B	32.1–72.0	46.9	42.5
Cd	5.70–1500	185	11.2
Co	6.17–24.9	13.9	9.68
Cr	150–3990	1560	1270
Cu	390–7140	1480	820
Fe	6920–48100	11760	13500
Hg	1.93–49.0	13.6	9.82
Mn	200–9870	1430	450
Mo	6.60–45.5	18.5	11.1
Ni	20–500	150	80
Pb	40–7480	1110	150
Se	4.23–17.2	8.51	7.02
Zn	740–45800	7220	1770
pH	6.3–8.3	7.58	7.50

METHODS AND MATERIALS

Bulk samples of soil (0–15 cm depth) were collected from 11 sites (Table 1) in the U.S. and transported to the Environmental Research Center, U.S. EPA, Cincinnati. At each site, the soils were sampled from an area that had no previous sludge application history (controls) and an adjacent area with similar soil properties that had sludge applied (treated), for a total of 22 soil samples. For the Blount soil from Indiana, a third soil was obtained by combining the treated and control soils (Blount 50/50). Thus, 23 soils were used in the experiment. Table 2 gives the chemical composition [14] of the sludge for all sites. For brevity, only the range, mean and median values are listed. Table 3 gives a comparison of some selected parameters for the sampling sites.

TABLE 3

Selected parameters of the sludges used at the 11 sites

Soil series	Reported years of sludge spreading*	Annual sludge application rate (dry m tons ha^{-1})	Cumulative total sludge (dry m tons ha^{-1})	Sludge digestion process
Congaree	11	28	308	Anaerobic
Salinas	7	21	147	Aerobic
Britwater	15	16	240	Aerobic
Burkhardt	6	16	96	Anaerobic
Pembroke	9	22	198	Anaerobic
Russell	12	30	360	Anaerobic
Morley	13	17	256	Anaerobic
Ross	5	65	325	Anaerobic
Del Rey	5	40	200	Anaerobic
Wea	30	65	1950	Anaerobic
Blount	10	11	110	Anaerobic

*Up to December 31, 1975.

A 10-kg batch of each air-dried soil was moistened and mixed in a twin-shell blender with 0, 5 or 10 μg Cd (as $CdSO_4$) solution g^{-1} soil. Additional Cd was, of course, present in the soil from previous sludge application. In addition, sufficient (0.5 g kg^{-1}) 15-30-15 fertilizer was added to the amended soils to meet the NPK requirements of the test crops. Although the pH values of the soils

TABLE 4

Selected properties of the soils used in the experiments

Soil series	Sludge treatment	Saturated soil pH		Total ($\mu g\ g^{-1}$)			Organic carbon (%)	CEC (meq/100 g)	Particle size (%)			Textural designation
		− Lime	+ Lime	Cd	Zn	Mn			Sand	Silt	Clay	
Congaree	−	3.9	7.0	0.3	37	385	1.3	6	65	28	7	Sandy loam
Congaree	+	4.1	5.8	4.6	611	107	23.4	22	70	29	1	Sandy loam
Salinas	−	6.8	7.3	4.2	100	427	2.7	29	26	39	35	Clay loam
Salinas	+	5.8	7.2	4.7	143	456	3.0	32	23	43	34	Clay loam
Blount (50/50)	−	6.5	7.4	0.4	71	1060	2.2	16	27	49	24	Loam
Blount (50/50)	+	7.1	7.2	18.4	306	1021	4.2	27	26	52	22	Silt loam
Britwater	−	6.0	7.2	0.5	48	1760	2.7	15	8	67	25	Silt loam
Britwater	+	6.3	7.1	0.7	120	1618	2.1	19	12	68	20	Silt loam
Burkhardt	−	5.5	7.2	0.4	53	498	1.5	19	77	20	3	Loamy sand
Burkhardt	+	5.9	7.1	0.4	56	470	2.1	15	89	7	4	Sand
Pembroke	−	6.3	7.3	0.6	84	1063	1.8	11	6	67	27	Silt loam
Pembroke	+	5.9	7.3	1.1	82	1256	2.3	14	16	62	22	Silt loam
Russell	−	6.1	7.3	0.5	60	1512	1.9	12	15	70	15	Silt loam
Russell	+	6.4	7.2	13.3	95	1134	3.2	14	13	69	18	Silt loam
Morley	−	6.2	7.3	0.5	74	534	2.9	13	40	41	19	Loam
Morley	+	6.1	7.3	0.9	142	693	2.3	14	31	40	29	Clay loam
Ross	−	6.4	7.3	0.8	100	1049	2.8	15	22	44	34	Clay loam
Ross	+	6.3	6.7	3.4	379	948	6.8	27	54	35	11	Sandy loam
Del Rey	−	6.9	7.4	0.7	79	460	4.8	22	26	49	25	Silt loam
Del Rey	+	7.0	7.0	6.3	497	410	7.9	30	34	46	20	Loam
Wea	−	6.3	7.3	1.1	111	1023	4.0	19	34	49	17	Loam
Wea	+	7.1	7.1	6.0	298	613	3.2	17	37	44	19	Loam
Blount	−	6.5	7.4	0.4	71	1060	2.6	15	27	49	24	Loam
Blount	+	7.1	7.1	41.0	564	1088	3.5	23	24	55	21	Silt loam

decreased following the addition of the fertilizer, they stabilized at the values presented in Table 4 prior to planting the first "Fordhook Giant" Swiss chard crop and remained constant prior to the addition of 1% $CaCO_3$. After mixing, 3.3-kg subsamples of the prepared soils were placed into 3.0-liter plastic pots lined with water-tight plastic bags and arranged in the growth chamber in a randomized complete block design, replicated three times. Swiss chard or corn (FRMO17 × H98 single cross) seed was placed directly into 2-cm pressed vermiculite cubes. Two weeks after planting and approximately 3 weeks after amending the soils with $CdSO_4$, two or four seedlings were transplanted into each pot. The plants were grown for 5 weeks in an environmental growth chamber maintained at 24°C during the day and 18°C at night with a 16-h photoperiod. Tops were then clipped 2 cm above the soil surface, rinsed in deionized water to remove soil particles and placed in paper bags. The plants were dried at 70°C and weights were recorded. Plant tops were then ground in a stainless steel Wiley mill and stored in glass vials for chemical analysis. Plant roots were removed from the soils, and the soils were air-dried, sieved, remixed, sampled and potted for planting of a second crop. The soil samples were taken to the laboratory for chemical analysis. This procedure was carried out for four successive crops (three Swiss chard and one corn). After the four crops, all soils were limed with 1% $CaCO_3$, repotted, and seeded to corn and then to Swiss chard, so that a total of six crops were grown sequentially in the treated soils.

Subsamples of the 23 soils were analyzed for selected chemical and physical properties. Soil pH of the saturated paste [15] was determined. A wet oxidation method [16] was used for organic carbon. The cation exchange capacity was measured using a technique developed by the U.S. Salinity Laboratory [15]. Mechanical analysis was performed by the hydrometer method [17]. The 500-mg dried and ground (60 mesh) soil samples were digested in 4 ml of 2:1 nitric–perchloric acid mix, brought to a 50-ml volume and stored in plastic bottles prior to analysis for Cd, Zn and Mn. Depending upon concentration, either flame or flameless atomic absorption procedures [18] were used for the analysis. The instrument used was equipped with a deuterium-arc background corrector. Standards and blanks were prepared to have the same acid concentration as the unknown samples. Details of chemical properties and particle size are given in Table 4.

Samples (250 mg) of the dried and ground plant material were digested in 2 ml of 2:1 nitric-perchloric acid mix, brought to a 50-ml volume with deionized water and analyzed for Cd [18]. Depending upon concentration, Cd was analyzed by either flame or flameless atomic absorption spectrophotometry with a deuterium-arc background corrector.

Yield and plant analysis data were subjected to a factorial type analysis of variance for a repeated measures experiment. The main plots of the experiment were a 12 × 3 × 2 randomized complete block design with three replications. In the analysis of variance the main plot effects are soils, sludge and Cd added and all interactions among these factors. In the significance tests for the liming effects, conservative tests using minimum degrees of freedom for the required F values for significance were utilized.

TABLE 5

Mean dry weight (g pot^{-1}) of Swiss Chard in relation to soil, cadmium added, sludge and lime regime

Treatment means	Unlimed			Limed		
	− Sludge	+ Sludge	Soil means[a]	− Sludge	+ Sludge	Soil means
Soil						
Congaree	3.99	3.99	3.99 e	3.40	4.47	3.94 de
Salinas	5.25	7.65	6.45 a	5.25	6.86	6.06 ab
Blount (50/50)	4.43	5.16	4.80 cd	4.92	5.93	5.43 bc
Britwater	4.78	4.84	4.81 cd	4.28	6.34	5.31 bc
Burkhardt	3.78	3.59	3.69 ef	4.71	4.59	4.65 cd
Pembroke	5.49	5.07	5.28 bc	6.18	6.11	6.15 ab
Russell	4.60	5.72	5.16 bcd	5.95	5.85	5.90 ab
Morley	5.51	5.49	5.50 b	5.69	5.86	5.78 ab
Ross	4.66	2.26	3.46 b	4.24	1.94	3.09 e
Del Rey	6.14	6.42	6.28 a	5.69	6.12	5.91 ab
Wea	6.14	4.68	5.41 b	6.06	6.92	6.49 a
Blount	4.43	4.92	4.68 d	4.92	6.39	5.66 ab
Cadmium added[a] (μg g^{-1})						
0	4.86 a	4.89 a		5.28 a	5.50 a	
5	5.03 a	5.13 a		4.90 a	5.54 a	
10	4.92 a	4.93 a		5.14 a	5.81 a	
Sludge	4.94 A	4.98 A		5.11 B	5.62 A	
Lime			4.96 B			5.37 A

[a] Means in a column (soil or cadmium added) followed by the same lower case letter or in a row (sludge or lime) followed by the same uppercase letter are not significantly different at the 0.05 probability level by Duncan's new multiple range test.

RESULTS AND DISCUSSION

Shoot weight relationships

The added Cd did not affect the yield of Swiss chard grown on the unlimed nor limed soils (Table 5). A significant increase in yield was evident on sludge-amended soils that were limed compared with the non sludge-amended soils, and a significant increase occurred for most limed soils compared with unlimed soils. The data also show that Swiss chard yield was significantly influenced by type of soil, which was probably due to differences in soil pH and other soil properties, such as organic matter, CEC, clay content, sludge properties, and fertility of the soils.

Significant main effects on total shoot weights of corn were found for all parameters except sludge for unlimed soils (Table 6). The 5 and 10 μg Cd g^{-1} soil addition significantly decreased the yield of corn compared to the 0 μg Cd g^{-1} addition rate. In general, corn yields were decreased by each increment of added Cd, except that there was no significant difference between the 5- and

TABLE 6

Mean dry weight (g pot $^{-1}$) of corn in relation to soil, cadmium added, sludge and lime regime

Treatment means	Unlimed			Limed		
	– Sludge	+ Sludge	Soil means[a]	– Sludge	+ Sludge	Soil means
Soil						
Congaree	2.44	4.05	3.25 b	4.83	6.19	5.51 a
Salinas	6.67	6.70	6.69 a	4.06	6.74	5.40 a
Blount (50/50)	4.77	5.57	5.17 d	4.98	3.51	4.25 cd
Britwater	6.21	6.01	6.11 b	3.97	4.65	4.31 c
Burkhardt	4.35	3.66	4.01 e	4.72	4.79	4.76
Pembroke	5.99	5.61	5.80 bc	6.08	4.94	5.51 a
Russell	6.22	5.31	5.77 bc	4.62	3.17	3.90 d
Morley	5.48	4.97	5.23 cd	4.87	4.38	4.63 bc
Ross	5.44	5.35	5.40 cd	4.66	4.14	4.40 bc
Del Rey	5.44	5.17	5.31 cd	4.83	3.84	4.34 c
Wea	6.07	4.73	5.40 cd	5.27	3.44	4.36 bc
Cadmium added (μg g^{-1})						
0	5.71 a	5.23 a		5.20 a	5.04 a	
5	5.33 b	4.99 b		4.74 b	4.39 b	
10	4.92 c	5.04 b		4.53 c	4.07 c	
Sludge	5.32 A	5.09 A		4.82 A	4.50 B	
Lime			5.20 A			4.66 B

[a] Means in a column (soil or cadmium added) followed by the same lowercase letter or in a row (sludge or lime) followed by the same uppercase letter are not significantly different at the 0.05 probability level by Duncan's new multiple range test.

10-μg Cd addition with the soils which had sludge applied to them but were not limed. For the unlimed soils, no significant differences occurred between yields of corn grown on sludge or nonsludge-amended soils, although there was a trend for sludge addition to suppress yields. There was a significant decrease in yields in the corn plants grown on the sludge-amended soils compared with the non-sludge-amended soils that had been limed. In contrast to the Swiss chard yields, liming the soils resulted in a significant decrease in corn yield compared with that of the unlimed soils. This decrease may be related to the availability of nutrients or to the Cd being more toxic to the corn plants growing on limed soils [1–3].

Plant composition

The only main effects that significantly influenced Cd concentration in chard and corn plants (Tables 7–10) were soils and Cd added. There was no significant sludge main effect on Cd concentration in the crops except on corn plants grown on limed soil. All interactive effects (S × SL, S × C, C × SL and S × SL × C) were also highly significant. However, for the soils where no

TABLE 7

Mean Cd concentration ($\mu g\ g^{-1}$) of Swiss chard plants on unlimed soil, in relation to Cd addition rate and sludge

Soil series	− Sludge			Soil means*	+ Sludge			Soil means
	Cd added ($\mu g\ g^{-1}$)				Cd added ($\mu g\ g^{-1}$)			
	0	5	10		0	5	10	
Congaree	0.9	51.1	118.0	56.7 a	9.9	36.8	64.8	37.2 d
Salinas	11.9	47.6	60.1	39.9 c	14.5	54.5	90.8	53.3 c
Blount (50/50)	2.0	26.1	44.4	24.2 fg	13.7	22.0	26.6	20.8 e
Britwater	0.8	36.1	57.5	31.5 d	1.0	15.5	29.0	15.2 f
Burkhardt	0.9	37.5	75.2	37.9 c	1.5	43.5	62.3	35.8 d
Pembroke	1.8	29.3	57.7	29.6 de	0.9	88.2	143.0	77.4 a
Russell	2.2	43.3	101.0	48.8 b	55.4	68.3	88.5	70.7 b
Morley	0.9	27.8	57.6	28.8 def	1.5	16.0	27.7	15.1 f
Ross	0.9	22.4	39.8	21.0 g	3.6	8.1	15.9	9.2 g
Del Rey	0.9	29.3	48.2	26.1 efg	6.2	10.8	17.5	11.5 fg
Wea	1.4	15.8	29.8	15.7 h	8.3	15.0	22.5	15.3 f
Blount	1.5	26.1	43.2	23.6 fg	17.5	20.5	29.4	22.5 e
Main effects								
Cd added	2.2 C	32.7 B	61.3 A		11.2 C	33.3 B	51.5 A	
Sludge				32.1 A				32.0 A
Mean for unlimed soils								32.1

*Means in a column followed by the same lowercase letter or means in a row followed by same uppercase letter under − sludge or + sludge are not significantly different at the 0.05 probability level by Duncan's new multiple range test.

additional Cd was added, there was a positive response to the previously added sludge Cd. Where additional Cd was added at a rate of $10\ \mu g\ Cd\ g^{-1}$ soil, there was less response to the added Cd in the sludge-amended soils than in the control soils. In most instances, the plant Cd content for the added $10\ \mu g\ Cd\ g^{-1}$ soil was less in those soils which had previously received sludge.

The coefficient of variation for the Cd content of chard and corn ranged from 10 to 12.6% and 16.2 to 21.7%, respectively. The range of Cd concentrations of shoots for the lowest Cd soil treatments ($0\ \mu g\ Cd\ g^{-1}$ added—minus sludge treatments, limed lowest, unlimed highest), excluding the Salinas soil series, varied as follows: 0.6–2.2 $\mu g\ Cd\ g^{-1}$, chard; 0.7–5.7 $\mu g\ Cd\ g^{-1}$, corn. The Cd concentration found in the control soils, except for the Salinas soil, are concentrations that one would expect to find in plants growing on non Cd-contaminated soils. The Salinas soil series is from an area in California that has high natural levels of Cd in the soil so that one would expect to find elevated Cd concentrations in plants grown on this soil. For the highest soil Cd treatment ($10\ \mu g\ Cd\ g^{-1}$), on average, chard and corn accumulated 41.4 and 83.8 $\mu g\ Cd\ g^{-1}$, respectively.

TABLE 8

Mean Cd concentration ($\mu g\, g^{-1}$) in Swiss chard plants on limed soil in relation to Cd addition rate and sludge

Soil series	− Sludge			Soil means	+ Sludge			Soil means
	Cd added ($\mu g\, g^{-1}$)				Cd added ($\mu g\, g^{-1}$)			
	0	5	10		0	5	10	
Congaree	1.0	27.2	56.9	28.4 a	1.0	10.0	17.5	9.5 de
Salinas	10.5	23.1	36.2	23.3 b	7.3	15.7	24.8	15.9 c
Blount	1.5	15.2	26.0	14.2 c	14.0	21.6	24.9	20.2 c
Britwater	0.6	13.2	21.5	11.8 cd	0.6	6.1	14.4	7.0 de
Burkhardt	0.6	18.1	40.8	19.8 b	0.9	28.8	41.8	23.8 b
Pembroke	1.0	8.1	15.1	8.1 d	0.7	11.7	14.8	9.1 de
Russell	0.7	11.4	26.4	12.8 cd	22.7	27.6	38.3	29.5 a
Morley	1.0	16.1	42.4	19.8 b	0.7	10.3	21.1	10.7 d
Ross	0.7	14.0	24.5	13.1 cd	1.9	5.7	6.6	4.7 e
Del Rey	0.7	16.0	21.4	12.7 cd	4.2	7.8	14.4	8.8 de
Wea	1.5	10.1	24.0	11.9 cd	8.5	15.9	26.2	16.9 c
Blount	0.6	14.7	24.3	13.2 cd	14.4	19.7	26.5	20.2 bc
Main effects								
Cd added	1.7 C	15.6 B	30.0 A		8.4 C	15.1 B	22.6 A	
Sludge				15.8 A				15.3
Mean for limed soils								15.3

[a] Means in a column followed by the same lowercase letter or means in a row followed by same uppercase letter under − sludge or + sludge are not significantly different at the 0.05 probability level by Duncan's new multiple range test.

The highest Cd concentrations in both species were found in those plants grown on the unlimed soils. Also, differences in uptake of Cd were evident between the two plant species. For example, in the unlimed soils which had sludge applied, chard plants accumulated between 15.9 and 143 $\mu g\, Cd\, g^{-1}$ plant material at the highest soil Cd addition, whereas corn accumulated between 34.0 and 155 $\mu g\, Cd\, g^{-1}$. In contrast to these elevated concentrations, shoot Cd concentrations for chard grown on the limed soils with added sludge varied from 6.6 to 41.8 $\mu g\, Cd\, g^{-1}$, while corn accumulated between 21.0 and 91.0 $\mu g\, Cd\, g^{-1}$. Thus, at least in the case of this single cross corn, the general observation that leafy vegetables are more responsive to soil Cd does not hold.

Studies of the effect of liming soils on Cd accumulation by different plants have almost always found lower plant accumulation from soils which contain lime compared with moderately to strongly acid soils [1–5, 19]. The authors suggest that the difference in Cd accumulation by plants grown on limed and unlimed soils is due to changes in solubility of Cd and physiological changes in ion uptake and transport due to variation in soil pH. It may also be important to consider the soil pH immediately adjacent to plant roots (rhizocylinder) rather than the pH of the total soil. This concept can be gleaned from the work

TABLE 9

Mean Cd concentration ($\mu g\ g^{-1}$) of corn plants on unlimed soil in relation to Cd addition and sludge

Soil series	− Sludge			Soil means[a]	+ Sludge			Soil means
	Cd added ($\mu g\ g^{-1}$)				Cd added ($\mu g\ g^{-1}$)			
	0	5	10		0	5	10	
Congaree	2.0	108.0	156.0	88.7 b	15.7	37.5	66.4	39.9 ef
Salinas	48.9	109.0	135.0	97.6 a	55.6	113.0	137.0	101.9 a
Blount (50/50)	3.5	81.8	113.0	66.1 de	58.6	72.1	79.3	70.0 c
Britwater	1.1	54.0	92.1	49.1 f	2.1	35.6	59.7	32.5 f
Burkhardt	1.6	87.3	138.0	75.6 c	2.0	102.0	155.0	86.3 b
Pembroke	1.3	75.3	107.0	61.2 e	6.1	73.9	115.0	65.0 cd
Russell	2.3	72.3	116.0	63.5 de	98.6	130.0	133.0	80.5 b
Morley	4.4	106.0	145.0	85.1 b	5.5	67.4	107.0	60.0 d
Ross	2.6	61.1	95.5	53.1 f	5.9	20.4	34.0	20.1 g
Del Rey	3.1	85.7	126.0	71.6 cd	29.3	69.3	87.1	61.9 cd
Wea	5.7	48.1	90.2	48.0 f	20.7	45.7	63.9	43.4 e
Blount	3.6	82.3	112.0	66.0 de	72.1	89.5	98.4	86.7 b
Main effects								
Cd added	6.7 C	80.9 B	119.0 A		31.0 C	71.4 B	94.6 A	
Sludge				68.9 A				65.7 A
Mean for unlimed soils								67.2

[a] Means in a column followed by the same lowercase letter or means in a row followed by same uppercase letter under − sludge or + sludge are not significantly different at the 0.05 probability level by Duncan's new multiple range test.

of Williams and David [12], which showed that NH_4 fertilizer increased Cd accumulation by plants. Smiley [20] showed that the form of N absorbed by the roots affected whether rhizocylinder soil pH declines (NH_4-N) or increases (NO_3-N). Thus, the implication of the Williams and David [12] data is that rhizocylinder pH affects plant accumulation of Cd.

Relationships between plant Cd and annual or cumulative additions of Cd

The effect of soils, sludge and lime on Cd uptake can be further evaluated from Tables 11 and 12, which show the increase of Cd in the plants (μg/g dry weight) per unit of Cd added to the soil (0, 5, and 10 $\mu g\ Cd\ g^{-1}$ soil treatment). Cadium increase was less for the soils with sludge added than for the soils with no sludge added. There was clear evidence of soil pH effect; lower pH soils generally showed a greater response. Evidently, after an application of sewage sludge the response to added Cd is decreased. Addition of lime to the soils for pH adjustment attenuated the response, but soils which had previously received sludge still showed a greater decrease than those which had not. It is

TABLE 10

Mean Cd concentration ($\mu g\ g^{-1}$) of corn plants on limed soil in relation to Cd addition and sludge

Soil series	− Sludge			Soil means[a]	+ Sludge			Soil means
	Cd added ($\mu g\ g^{-1}$)				Cd added ($\mu g\ g^{-1}$)			
	0	5	10		0	5	10	
Congaree	0.9	75.7	115.0	63.9 a	14.7	25.7	48.6	29.7 cd
Salinas	23.8	58.0	78.0	53.3 b	26.6	55.3	79.3	53.7 b
Blount (50/50)	1.2	24.0	41.3	22.2 fg	51.3	66.0	72.0	63.1 a
Britwater	0.7	22.7	36.7	20.0 g	2.8	39.0	59.0	33.6 c
Burkhardt	1.0	39.0	80.0	40.0 c	2.4	60.0	91.0	51.1 b
Pembroke	2.1	28.1	41.0	23.7 efg	1.3	35.0	49.0	28.4 d
Russell	1.8	33.0	49.0	27.9 de	52.0	65.0	83.0	66.7 a
Morley	2.6	43.0	64.0	36.5 c	3.0	35.0	61.0	33.0 cd
Ross	2.1	28.0	46.0	25.4 ef	3.0	15.0	21.0	13.0 e
Del Rey	2.2	32.7	56.0	30.3 d	24.0	54.0	77.0	51.7 b
Wea	3.2	23.0	43.0	23.1 efg	16.0	31.0	43.0	30.0 cd
Blount	1.8	29.0	45.0	25.3 ef	59.0	65.0	79.0	67.7 a
Main effects								
Cd added	3.6 C	36.4 B	57.9 A		21.3 C	45.5 B	63.6 A	
Sludge				32.6 B				43.5 A
Mean for limed soils								38.1

[a] Means in a column followed by the same lowercase letter or means in a row followed by same uppercase letter under − sludge or + sludge are not significantly different at the 0.05 probability level by Duncan's new multiple range test.

TABLE 11

Regression coefficients (b)[a] calculated by linear regression analysis of Swiss chard plant Cd in relation to Cd added to soil. Data is calculated from Tables 7 and 8

Soil series	Unlimed		Limed	
	− Sludge	+ Sludge	− Sludge	+ Sludge
Congaree	11.7	5.5	5.6	1.7
Salinas	4.8	7.6	2.6	1.8
Blount (50/50)	4.2	1.3	2.5	1.1
Britwater	5.7	2.8	2.1	1.4
Burkhardt	7.4	6.1	4.0	4.1
Pembroke	14.2	5.6	1.4	1.4
Russell	9.9	3.3	2.6	1.6
Morley	5.7	2.6	4.1	2.0
Ross	3.9	1.2	2.4	0.5
Del Rey	4.7	1.1	2.1	1.0
Wea	2.8	1.4	2.3	1.8
Blount	4.2	1.2	2.4	1.2
Mean	6.6	3.3	2.8	1.6

[a] $y = a + b(x)$ where y = plant Cd, b = slope and x = Cd added.

TABLE 12

Regression coefficients (b)* calculated by linear regression analysis of corn plant Cd in relation to Cd added to soil. Data is calculated from Tables 9 and 10

Soil series	Unlimed		Limed	
	– Sludge	+ Sludge	– Sludge	+ Sludge
Congaree	15.4	5.1	11.4	3.4
Salinas	8.6	8.1	5.4	5.3
Blount (50/50)	11.0	2.1	4.0	2.1
Britwater	9.1	5.8	3.6	5.6
Burkhardt	13.6	15.3	7.9	8.9
Pembroke	10.6	10.9	3.9	4.8
Russell	11.4	3.4	4.7	3.1
Morley	14.1	10.2	6.1	5.8
Ross	9.3	2.8	4.4	1.8
Del Rey	12.3	5.8	5.4	5.3
Wea	8.5	4.3	4.0	2.7
Blount	10.8	2.6	4.3	2.0
Mean	11.2	6.4	5.4	4.2

*$y = a + b(x)$ where y = plant Cd, b = slope and x = Cd added.

also apparent from a comparison of the O-Cd-added rate with one another (Tables 7–10), that the Cd which had previously been added to the soil was still available. Thus, both annual and cumulative additions are important. The importance of both annual and cumulative rates of Cd addition can be seen in the Blount soil series. In the case of the Blount soil series, a 50-50 mixture of control and treated soil was utilized as well as the control and treated soils. This allows for the development of the relationship between Cd added from sludge and plant Cd as well as response to added inorganic Cd (Fig. 1). These data indicate that the relationship between crop Cd and soil Cd depends upon the level of Cd originally present in the soil. For example, a regression analysis of the 0 μg Cd g^{-1} addition shows a cumulative effect ($y = 2.7 + 0.35x$, $r = 0.9974^{**}$; y = plant Cd concentration, μg g^{-1}; x = soil Cd concentration, μg g^{-1}). However, as inorganic Cd is added to each of these soils, it is more available than that which is cumulative, but its availability decreases as the cumulative amount present in the soil initially increases. If the same regression analysis is used at the 5 μg Cd g^{-1} annual inorganic-Cd-addition treatment, the cumulative response curve has a negative slope ($y = 27.66 - 0.16x$, $r = 0.9994^{**}$). This implies that the more Cd added, the lower the concentration in the plant and that both annual and cumulative Cd additions are important to regulating plant uptake of Cd. This difference could also be attributed to the source of Cd (sludge Cd vs. $CdSO_4$-Cd). Data from Illinois [21] also indicate that the slopes from the annual Cd additions are greater than those from the cumulative Cd additions. Their data imply that the Cd was becoming less available with time and that both annual and cumulative additions are important. It becomes apparent that for long-term response, the

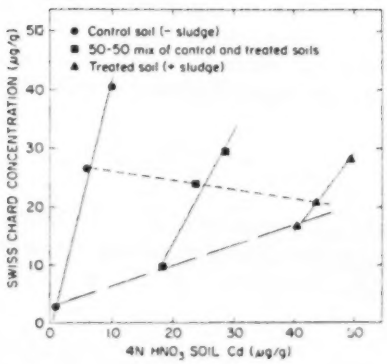

Fig. 1. Effects of mixing control and treated soils on the relationship between plant Cd and soil Cd.

increase in crop Cd will be less than has been shown with addition of recent Cd. Thus, both annual and cumulative additions need to be considered.

The CAST Reports [22, 23] also indicate that the effects of Cd application depend upon the mode of application. For example, single application vs. repeated applications may produce different results although the cumulative amount of Cd applied is equal. This concept is supported by the work of Hinesly et al. [21, 24], Giordano and Mays [25] and Bates et al. [26]. Their data imply that there is not a single linear relationship between total soil Cd and plant Cd, but that the relationship is a function of the annual application rate. Hinesly et al. [21, 27] stress the importance of the role of annual Cd application, compared with cumulative Cd application, suggesting that only the current year's Cd application is important. If so, the previously applied Cd must be nearly unavailable to the plant, which is certainly not the case with the present data.

Presently, we are unable to account for these differences in plant response to annual and/or cumulative application rates of Cd. This causes a dilemma in that limits to soil application based on accumulation of Cd encourage small applications to new areas of land, whereas an annual application limit encourages application to the same land for extended periods. The question raised is whether the objectives are to minimize the fraction of the soil-applied Cd which is taken up by the crop and thus the amount of Cd exposure from food for the total population, or to limit the amount of Cd ingested by any individual to some acceptable level, thus creating a smaller potential exposure for more people. If a low limit is placed on the total amount of Cd which can be applied to soil, then, at some time, all available soils will have accumulated that level of Cd. Thus, if there were no reduction in plant availability of Cd over time, all crops would be at the higher Cd content, and the exposure of the total population would be increased. Conversely, if a limit were placed on the annual amount of Cd which could be applied to soil, and the same land area is used continually, the Cd content of only a small fraction of all crops would be

increased. However, the increase in Cd content is dependent upon cumulative loading rather than annual loading. Thus, a consideration of both annual and cumulative Cd additions would be necessary to predict plant Cd concentration when annual applications of Cd occur. After termination of application and establishment of equilibrium (steady state) conditions, only the cumulative response would be important.

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